Metal-loaded Liquid Scintillator

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The Trend of Neutrino Detector

1961

Radiochemical

Water

HOMESTAKE Radiochemical Detector

615 tons of C_2Cl_4 ; ${}^{37}Cl + v_e \rightarrow {}^{37}Ar + e^- (\sim 40 \text{ years})$

GALLEX Radiochemical Detector

30 tons of Ga; $^{71}Ga + v_{\rho} \rightarrow ^{71}Ge + e^{-}$ (1986 - 1998)

SNO Water Čerenkov Detector (SNOLab)

1-k tons of ultra-pure D_2O (1996 - 2006)

Long-Baseline Neutrino Experiment (LBNE)

 V_u beam from accelerator to DUSEL (began~2002)

High-Precision Reactor θ_{13} Real-time Detector (Project)

200 tons of 0.1% Gd in Liquid Scintillator (began ~2003)

Liquid SNO+ Neutrinoless Double Beta Decay Real-time Detector (R&D) **Scintillator**

1-k tons of 0.1% Nd in Liquid Scintillator (began ~2005)

LENS Real-time LS Detector (R&D)

100 tons of ~8% ¹¹⁵In in Liquid Scintillator (began ~2000)

Neutrino Application

- Geo-neutrino and/or Nonproliferation and/or Reactor Monitoring
- Li/Ca-loaded Liquid Scintillator (began 2008)

What do we need from M-LS?

 Preparation of high-quality M-LS must be Reliable and Reproducible: long-term stability and long attenuation length with suitable photon-production

At BNL:

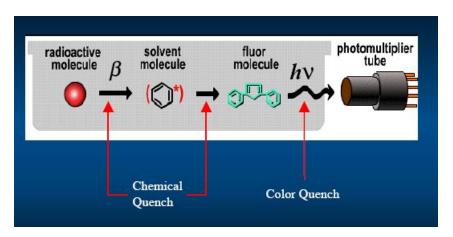
- Solvent-extract organometallic compound into LS with carboxylic acid, RCOOH
- Study effects of inclusion in organic phase of OH-, H₂O, free carboxylic acid, CI-
- Characterize how parameters affect light yield, chemical stability, final metal ions dissolved in LS, and chemical composition
- ➤ Develop a variety of chemical techniques for chemical/radioactive purifications of all starting materials, which is the key of clean M-LS
- Remove the U/Th/K from the starting materials to increase the sensitivity

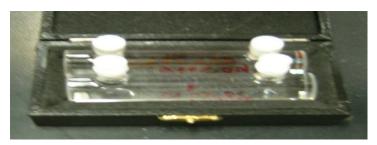
M-LS Criteria for Neutrino Experiments

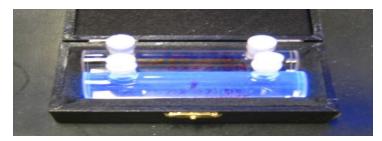
- ➤ Long-term Stability: > 3 years
- ➤ Long attenuation length: > 10 m
- ➤ High photon yield
- > Low radioactive background
- > ESSH

Components of LS

- **Aromatic solvent** that contains a high density of π -electrons for energy transfer
- ❖ Fluor that transfers the energy (<400 nm) to light (>400 nm) within the optimal detection range of PMT







<u>Metal selection for different</u> <u>experiments</u>

- Nd for SNO+
 - Serve as a double-beta decay target
 - Large Q(3.37 MeV) and favorable matrix element
- Gd for Daya Bay
 - Enhance neutron-capture cross section
 - Shorten neutron capture time
 - A high release energy from capture, good for suppressing background
- In for LENS
 - Low threshold
 - Directly measures neutrino energy
 - Triple tag effect

Selection of Liquid Scintillator

- High density, flash point, low toxicity, and low cost
- Chemical compatibility
- High light yield and long attenuation
- Able to load organometallic compound

Study of Liquid Scintillator

	Gd Loading	d (g/cm³)	UV Abs ⁴³⁰ before / after	Abs_{260}	n ²⁰	Light Yield	H atoms‡ per c.c	Flash Point
PC	Yes	0.889	0.008 / 0.002	2	1.504	1	5.35×10 ²²	48 C
РСН	Yes	0.95	0.072 / 0.001	1.7	1.526	0.46	5.71×10 ²²	99 C
DIN	Yes	0.96	0.040 / 0.023	>10	-	0.87	5.45×10 ²²	>140 C
PXE	Yes	0.985	0.044 / 0.022	2.1	-	0.87	5.08×10 ²²	167 C
LAB	Yes	0.86	0.001 / 0.000	1	1.482	0.98	6.31×10 ²²	130 C
Mineral Oil C ₂₄ ~C ₂₈	No	0.85	0.002 / 0.001	1	~1.46	~	6.73 - 8.00 $\times 10^{22}$	215 C
Dodecane	No (<20%)	0.75	0.001 / 0.000	1	1.422	~	6.89×10 ²²	71 C

M. Yeh et. al Nuclear Instruments and Methods in Physics Research Section A: Gadolinium-loaded liquid scintillator for high-precision measurements of antineutrino oscillations and the mixing angle, θ13 Volume 578, Issue 1, 21 July 2007, Pages 329-339

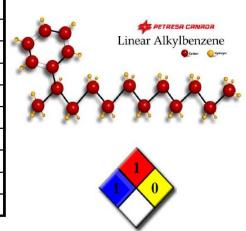
LAB Specification

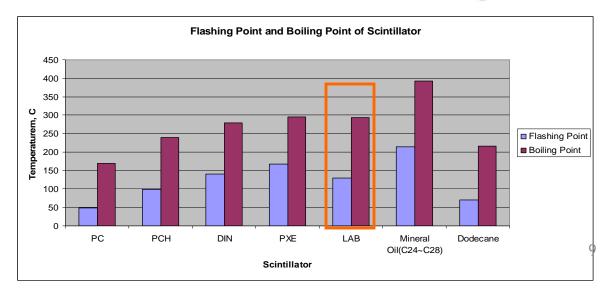
LAB is attractive:

- High flashpoint
- Biodegradable
- Millions of tons of it are produced annually for detergent industry

linear alkyl chains of 10-13 C atoms with a benzene ring; used primarily for the production of biodegradable synthetic detergent

Boiling Point (°C)	275 - 307	
Melting Point (°C)	< -50	
Flash Point (°C)	130	
Vapor Pressure (mmHg)	< 0.1 mmHg @ 20°C	
Vapor Density (Air =1)	8.1	₩
Solubility in Water	Insoluble	<i>6</i>
Molecule Weight	233 – 237 g/mol	
рН	Not applicable	
Viscosity	5 – 10 cps @ 20°C	
Evaporation Rate (water =1)	Not applicable	





Selection of Extractant

		-		
Extractant Gd form in LS		Comparison		
alcohols	GdCl₃ .6H₂O	GdCl ₃ quickly dissolved in the mixture of alcohol and PC; low light yield and not stable possibly due to quenching effect and high vapor pressure.		
P=O compounds	Gd(carboxylate) _x . (Cl) _y .(OH) _z .(H ₂ O) _a . (TOPO) _b	high extraction efficiency; moderate attenuation length; not stable for long term (phosphorus might interact with other compounds; CHOOZ experience of NO ₃)		
carboxylic	Gd(carboxylate) _x . (Cl) _y .(OH) _z .(H ₂ O) _a	high extraction efficiency; long attenuation length; high light yield; very stable (>14 months for Gd-LS and over 2.5 years for In-LS since synthesis)		

Selection of Carboxylic Complexing Ligands

CARBOXYLIC ACIDS FOR LENS YES

	CARBUATLIC ACIUS TUR LENS TES					
WATER SOLUBLE	$\begin{array}{c} 0 & 2C \\ \text{OH} & C - CH_3 \\ \text{ACETIC (AcA)[60]} \end{array}$	OH C CH ₃ PROPIONIC (PA)[74]	OH CH3 4C S CH3 OH CH3 ISOBUTYRIC (IBA)[88]	OH CC CH ₃ CH ₃ S BUTYRIC (BA)[88]		
	OCH3 CH3 OH ISOVALERIC (IVA)[102]	OH CH ₃ 5C OH CH ₃ CH ₃ TRIMETHYLACETIC (TMAA)[102]	OH METHYLBUTYRIC (MBA)[102]	0 5C S OH CH ₃ VALERIC (VA)[102]		
- WATER INSOLUBLE -	CH ₃ OH CCH ₃ CH ₃ CH ₃ CH ₃ CH ₃ CH ₃ CH ₃	CH ₃ 6C OH ETHYLBUTYRIC (EBA)[116]	CH ₃ CH ₃ CH ₃ CH ₃ 2,2DIMETHYLVALERIC (DMVA)[130]	OH CH3 OH CH3 HEXANOIC (HxA)[116]		
	CH ₃ 8C CH ₃ CH ₃ OH CH ₃ CH ₃ ETHYLHEXANOIC (EHA)[144]			OH CH3 CH3 CH3 OH CH3 CH3 TRIMETHYLHEXANOIC (TMHA)[158]		

Different carboxylic acids for different metal ions

Loading concentration

Loading efficiency

Light yield

Synthesis of M-LS

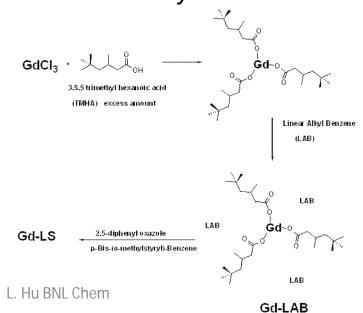
Solvent Extraction vs. Solid dissolution

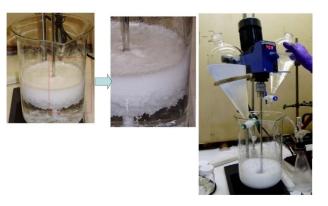
LAB + Carboxylic acid (in H_2O)+MCl₃ (in H_2O) Carboxylic Acid (in H_2O)+MCl₃ (in H_2O)





Remove water layer





Collect the solid
Wash
Dissolve the solid in LAB
Wait for it to get cleared
Leach out the additional water

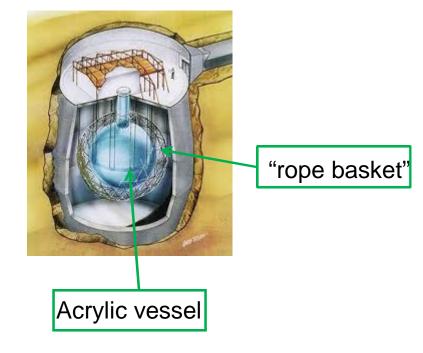
Nd-loaded LS for SNO+

1-k tons of 0.1% Nd in Liquid Scintillator

- ➤ Neutrinoless double beta decay (150Nd)
- ➤ Determine the mass/nature of the neutrino



SNO's ultra-pure water systems will be replaced by similar purification systems that can eliminate trace radioactivity from an organic liquid scintillator



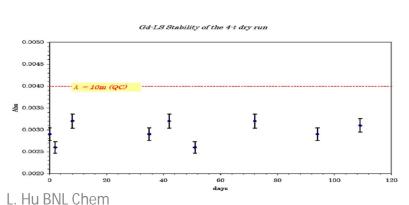
Gd-loaded LS for Daya Bay

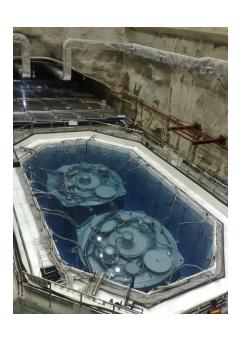
200 ton 0.1% gadolinium-loaded liquid scintillator (Gd-LS)

Measure reactor \overline{v} for θ_{13} at 1%

Gd-LS was produced in multiple batches but mixed in reservoir on-site, to ensure identical detectors.







Detectors in Hall 1

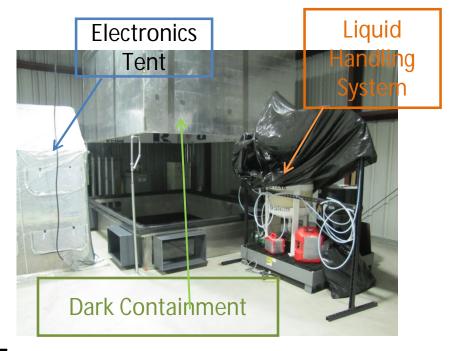
In-loaded LS for LENS

125 tons of ~8% ¹¹⁵In Liquid Scintillator for full scale LENS

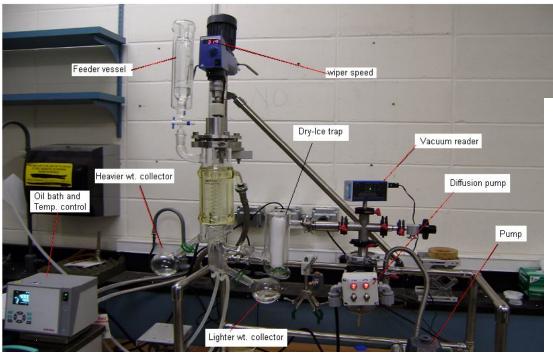
- > The real time primary low energy (sub-MeV) solar neutrino spectrum
- > Solar metallicity and luminosity



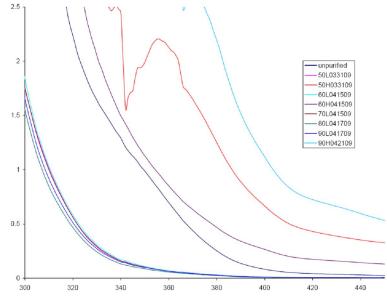
A view inside KURF from above microLENS liquid handling



Purification of Water and TMHA



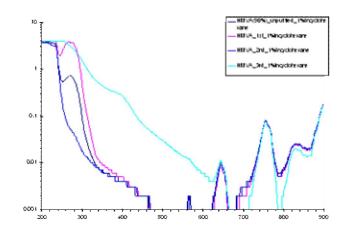




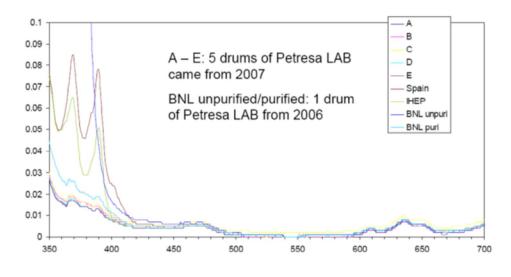
L. Hu BNL Chem

Purification of HMVA and LAB

- Automatic distillation of HMVA
- Dry column purify LAB



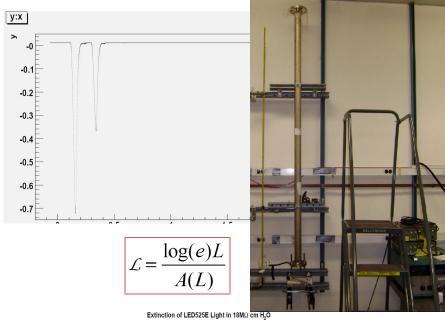


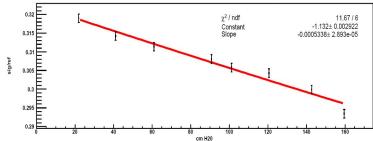




M-LS Characterization (I)

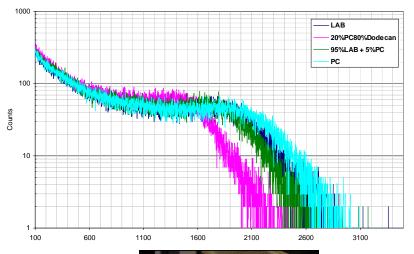
• $L_{1/e}$ (att. length) by 2-m dual-beam, vertical LED system.





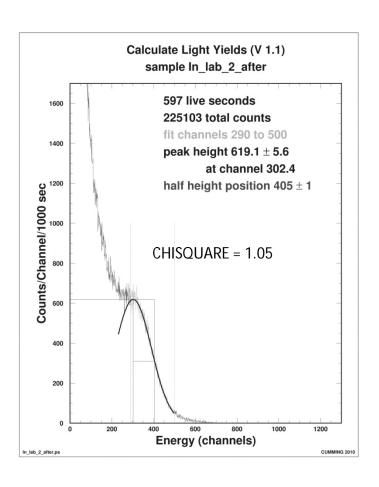
 18.9 ± 1.2 m for $18M\Omega$ H₂O at 525nm, <u>J. Goett</u>

• Light Yield (S%)



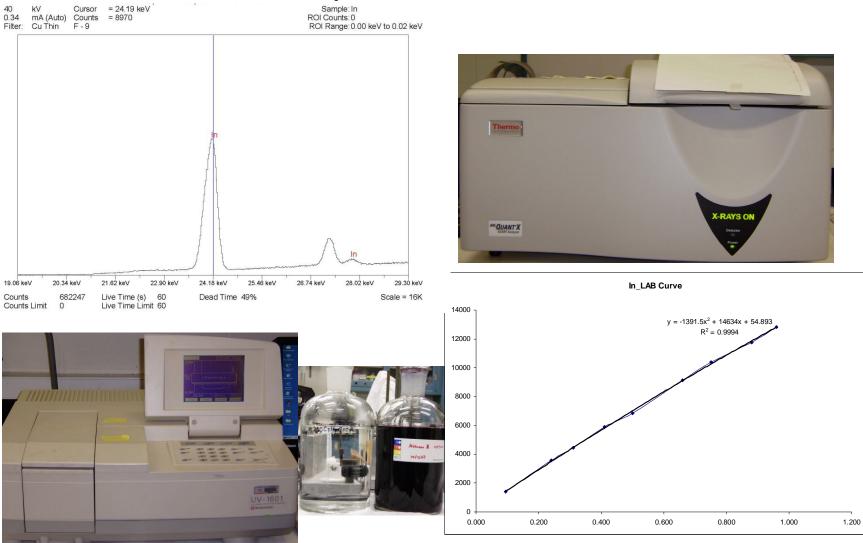


Light Yield Percentage Calculated Program



M-LS Characterization (II)

• [Mⁿ⁺] by XRF and UV



M-LS Characterization (III)



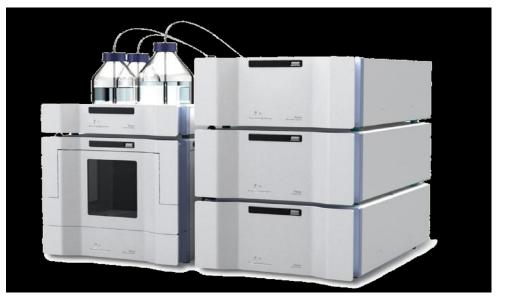


IR Qualification and quantification

Florescence Spectrometer Excitation, Emission, and lifetime of LS

M-LS Characterization (IV)





GC/MS Impurities of chemicals

LC/MS Materials compatibilities

<u>Summary</u>

- *M-loaded LS becomes one of main-stream detection mediums from the past 5 years.*
- BNL is one of the facility that has the expertise and equipments for all-aspect development of organometallic liquid scintillator including (1) synthesis, (2) characterization and (3) purification of M-LS.
- A variety of metallic ions has been loaded into liquid scintillator at different concentrations (0.1 10%) with <u>high-quality, large-scale production (> hundreds of tons) of M-LS</u> for different neutrino experiments
 - M-LS are stable for long period of time (>3.5 yrs for 10% In-LS; >2 yrs for 1% Nd-LS; and >3 yrs for 0.1% Gd-LS).
- M-LS use of veto or calibration are under development.

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